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Analytical Solution to Evaluate Salt Precipitation during CO_2 Injection in Saline Aquifers

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Abstract

The amount of salt precipitation and the region in which it occurs are important parameters in CO_2 storage management. For this study, an analytical model, as a simple and efficient tool to predict the amount of salt precipitation over time and space, is developed. The analytical model is then extended to evaluate the effect of salt precipitation on permeability in terms of a time-dependent skin factor. The analytical model is applied to a specific problem with available numerical solution, where a close agreement is observed. This is used to evaluate the effect of assumptions made in development of the analytical solution.

Keywords: CO_2 sequestration; salt precipitation; analytical solution

1. Introduction

As concern about the adverse consequences of anthropogenic climate change has grown, so too has research into methods to reduce the emissions of greenhouse gases. Carbon dioxide storage could enable the continued use of fossil fuels with reduced CO_2 emissions. Among various options of carbon dioxide storage, CO_2 can be sequestered in deep aquifers. The characteristics of a preferred storage site include (i) high capacity to allow storage of large volumes of CO_2, (ii) high injectivity to allow injection at desired rates and (iii) containment to ensure limited leakage (if any). Injectivity is not only a function of static reservoir properties (e.g. permeability and pay thickness) but also can change with time. This paper studies salt precipitation in the vicinity of the injection well, as one of the time-dependent phenomena that could affect injectivity. In particular an analytical model is developed to estimate the radius of the dry-out zone (where salt precipitation occurs) and the fraction of the pore volume that will be occupied by salt. This is achieved with the use of the theory developed by Orr [1] for linear two-component two-phase displacement. We redevelop this model for radial displacement of brine by CO_2. Although the flow is never one-dimensional in actual field-scale, the one-dimensional solution can describe the behaviour of CO_2 storage during the injection period where the flow is mainly along the reservoir bedding.

In the following paragraphs, the physical problem is described. This is followed by development of a mathematical model for radial CO_2/brine binary displacement. Then, the salt precipitation is formulated for the

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region where vaporization occurs in the binary system and included in the model. Next, salt precipitation effect on the permeability is studied and presented in terms of a time-dependent skin factor. Last, the analytical model is applied to a specific problem with an available numerical solution, to evaluate the effect of assumptions and approximations made in the development of the analytical solution.

2. Physical Problem

Consider 1-D CO_2 injection through a well located in the centre of an infinitely large saline aquifer. The following is the possible phase arrangement: deep in the reservoir, where the brine has not been in contact with the injected CO_2, only the fresh brine exists (region 1 in Figure 1). In the region behind the fresh brine, the brine contains the CO_2 component. In this region both aqueous and gaseous phases are mobile and in equilibrium (region 2 in Figure 1). The gaseous phase saturation of this region is increasing toward the injection well. The increase in gaseous phase saturation leads to excess water vaporization into the gaseous phase and, therefore, to higher brine salinities (since the solid salt in the aqueous phase remains constant while the water content reduces). This salinity increase causes the CO_2 dissolution to decrease also (salting out effect). Moving further toward the injection well, where the salinity level reaches that of halite-saturated brine, the solid salt precipitation begins. Thus a region of 3-phases (gaseous, aqueous and solid) and 3-components (water, CO_2 and salt) develops (region 3 in Figure 1). In this region the concentration of components in each phase is independent of total concentrations. The gaseous phase saturation increases toward the injection well while the aqueous phase saturation decreases. At certain saturation the aqueous phase flows slowly enough that it can be fully vaporized by the injected CO_2. This allows a 2-phases (solid and gaseous) and 3-components (water, CO_2, and salt) region to develop (region 4 in Figure 1). Further toward the injection well, the fresh injected CO_2 can transport the vaporised water outward. This allows development of a 2-phases (solid and gaseous) and 2-components (CO_2 and salt) region (region 5 in Figure 1) [2,3].

2-Phases, 2-Comps	2-Phases, 3-Comps	3-Phases, 3-Comps	2-Phases, 3-Comps	1-Phase, 2-Comps
Mobile gaseous (CO_2)	Mobile gaseous (CO_2, water)	Mobile gaseous (CO_2, water)	Mobile gaseous (CO_2, water)	
Solid(salt)	Solid(salt)	Solid (salt)		
		Mobile aqueous (CO_2, water, salt)	Mobile aqueous (CO_2, water, salt)	aqueous (water, salt)
5	**4**	**3**	**2**	**1**

Figure 1: Arrangement of different phases during 1-D CO_2/Brine displacement.

3. Analytical Model

Analytical modelling of the physical problem described above could consider a ternary system with three-phases (aqueous, gaseous, and solid) and three-components (CO_2, water, salt). The problem of three-phase compositional flow has been studied in several publications [4,5]. The full three-phase solutions are not simple to construct. The system studied in this work is a limiting case of the more general ternary problem where one of the phases is immobile (the solid salt). This leads to a simplified solution. In this paper the intent is not to solve the three-phase three-component problem directly. Rather, the system is approximated by a binary system with two-phases (aqueous and gaseous) and two-components (CO_2 and brine). In departure with the previous papers on the subject, volume change upon mixing is accounted for [6,7]. Then the amount of salt precipitation (solid phase saturation) is calculated separately based on the amount of vaporization. We then check the validity of this approximation by comparing the analytical results to the numerical ones for a specific case.

In the following, the analytical model for the binary system is presented. The solution includes two shocks that divides the medium into three regions. Far from the injection well -ahead of a leading shock- a fresh brine region exists that is being displaced by the injected CO_2. Upstream of the leading shock, a region of two-phase flow exists (equilibrium region), in which an aqueous phase (brine and dissolved CO_2) and a gaseous phase (CO_2 and vaporized

water) flow. A trailing evaporation shock separates the equilibrium region from the dry-out region closest to the wellbore where salt has precipitated. Therefore in this solution, the region arrangement reduces to that shown in Figure 2. The assumptions that lead to these three regions are discussed later in this paper.

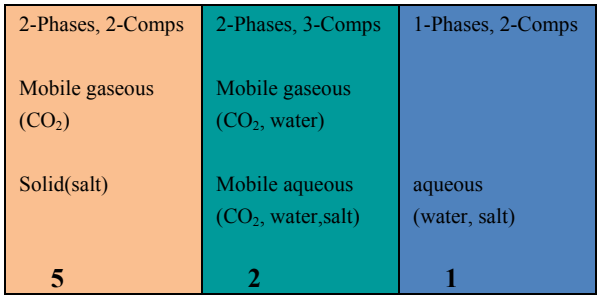

Figure 2: Arrangement of different phases during 1-D CO_2/Brine displacement based on the presented model in this paper.

After analytical modelling of the binary system, the amount of salt that precipitates behind the trailing shock is formulated to be included in the model. Salt can also be precipitated due to the reaction of the aquifer rock with involved fluids; however, the rock is assumed as non-reactive in this study.

4. Analytical Model for the Binary System

The solution in this section considers a binary system composed of two-phases (aqueous and gaseous) and two-components (CO_2 and brine). The components transfer between the displacing injected gas and displaced aquifer brine. Then the resulting aqueous phase includes brine with some CO_2. Similarly, the gaseous phase will only contain CO_2 and brine. However, the water properties are assigned to brine in the gaseous phase. The assumption here is that salt and water act together. With this assumption, the problem becomes similar to the gas/liquid displacement problem solved by many including Orr [1]. Application of this solution to a 1-D radial problem is given by Zeidouni et al. [8]. The model assumes homogeneity, no diffusion/dispersion, non-reactive rock, local phase equilibrium, and constant pressure and temperature. Starting with the continuity equation and using the fractional flow concept, the following first-order nonlinear hyperbolic partial differential equation is obtained:

$$\frac{\partial G_{CO2}}{\partial t}+\frac{q_{inj}}{\pi h\phi}\frac{dH_{CO2}}{dG_{CO2}}\frac{\partial G_{CO2}}{\partial r^2}=0 \qquad \textbf{(1)}$$

where r and t are the radial coordinate and time. G_{CO2} is total molar density (global concentration) of CO_2 component which is normalized with respect to the molar density of injected fluid. H_{CO2} is total molar rate of CO_2 component that is normalized with respect to molar injection rate. Exact definitions are:

$$G_{CO2}=\rho_{aD}\omega_{CO2,a}S_a+\rho_{gD}\omega_{CO2,g}S_g \quad \text{and} \quad H_{CO2}=\rho_{aD}\omega_{CO2,a}f_a q_D+\rho_{gD}\omega_{CO2,g}f_g q_D \qquad \textbf{(2)}$$

Similar equation can be written for brine component also. With the assumptions of no gravity and capillary forces the gaseous phase fractional flow is obtained by:

$$f_g=\frac{1}{1+\frac{k_{ra}}{k_{rg}}\frac{\mu_g}{\mu_a}} \qquad \textbf{(3)}$$

where k_{ra} and k_{rg} are the relative permeability of aqueous and gaseous phases respectively that depend only on gaseous phase saturation. μ_a and μ_g are the viscosity of aqueous and gaseous phases respectively. Aqueous phase fractional flow will be $1\text{-}f_g$.

The solution to Equation 1 is given by:

$$\left(\frac{r^2-r_w^{\,2}}{t}\right)_{G_{CO2}}=\frac{q_{inj}}{\pi h\phi}\left(\frac{dH_{CO2}}{dG_{CO2}}\right)_{G_{CO2}} \quad \text{or} \quad \left(\frac{r^2-r_w^{\,2}}{t}\right)_{G_{CO2}}=\frac{q_{inj}}{\pi h\phi}\left(q_D\frac{df_g}{dS_g}\right)_{G_{CO2}} \qquad \textbf{(4)}$$

where r_w is the wellbore radius, and $q_D=q_t/q_{inj}$. q_t is the total local flow rate defined as summation of the aqueous and

gaseous phase flow rates. In solution given by Equation 4, where mass transfer between phases occur and there is volume change upon mixing, q_t is not equal to q_{inj}, and varies across different regions. For any specific value of G_{CO2} this equation gives a specific value for the similarity variable (wave velocity) $\eta=(r^2-r_w^2)/t$ which is defined based on the dependent variables (r,t). This means that the problem has a similarity property, since the solution depends on radial distance r and time t only through the similarity variable η. The solution is expressed in terms of gaseous phase saturation or CO_2 overall mole fraction versus the similarity variable.

The solution given by direct use of Equation 4 is physically impossible because of multiple saturation values at a single location. Ignoring the capillary pressure (and also the diffusion process) causes this multi-valued solution. Two shocks (discontinuities in saturation and concentration) are introduced to resolve this inconsistency: trailing shock and leading sock. The trailing shock is the shock which connects the single-phase gaseous region to the two-phase region. The shock that connects the two-phase region to the single-phase aqueous region is called the leading shock, as it is always ahead of the trailing shock.

For the case of pure CO_2 injection in an aquifer that initially contains brine only(no initially dissolved CO_2), the placement of the shocks could be determined graphically. The method uses the gaseous phase fractional flow versus the gaseous phase saturation curve (f_g-S_g curve) to obtain the saturation at the trailing and leading shocks. The method is illustrated in figure 3. Gaseous phase saturation downstream of the trailing shock is the tangent point of the tangent line drawn from point J on the f_g-S_g curve. A tangent line drawn from point I on the f_g-S_g curve gives the gaseous phase saturation upstream of the leading shock. Both I and J points are located on a unit slope line drawn from the origin and can be obtained by:

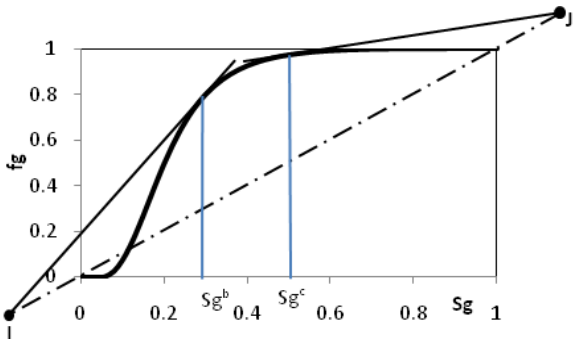

Figure 3: Graphical Method

$$\left(S_g^I, f_g^I\right)=\left(\frac{\rho_{aD}\omega_{CO2,a}}{\rho_{aD}\omega_{CO2,a}-\rho_{gD}\omega_{CO2,g}}, \frac{\rho_{aD}\omega_{CO2,a}}{\rho_{aD}\omega_{CO2,a}-\rho_{gD}\omega_{CO2,g}}\right) \quad \textbf{(5)}$$

$$\left(S_g^J, f_g^J\right)=\left(\frac{\rho_{aD}\omega_{Brine,a}}{\rho_{aD}\omega_{Brine,a}-\rho_{gD}\omega_{water,g}}, \frac{\rho_{aD}\omega_{Brine,a}}{\rho_{aD}\omega_{Brine,a}-\rho_{gD}\omega_{water,g}}\right) \quad \textbf{(6)}$$

To determine the similarity variable at the shock(based on Equation 4) the local flow rate at the two-phase region (q_D^c) is required which is obtained by:

$$q_D^c=\frac{(\rho_{aD}\omega_{CO2,a}-\rho_{gD}\omega_{CO2,g})S_g^c-(\rho_{aD}-\rho_{gD})S_g^c+(1-\omega_{CO2,a})\rho_{aD}}{(\rho_{aD}\omega_{CO2,a}-\rho_{gD}\omega_{CO2,g})f_g^c-(\rho_{aD}-\rho_{gD})f_g^c+(1-\omega_{CO2,a})\rho_{aD}+\rho_{aD}\rho_{gD}(\omega_{CO2,a}-\omega_{CO2,g})(S_g^c-f_g^c)} \quad \textbf{(7)}$$

For all the saturations between the trailing shock and leading shock saturations the similarity variable could be obtained based on equation 4. The details of derivation of the above graphical method are given by Zeidouni et. al. [8].

5. Salt Precipitation in the Dry-out Zone

In this section, the salt precipitation is modelled to be included in the radial CO_2/brine displacement that was modelled above. Salt precipitation may occur due to vaporization of brine into the gaseous phase during CO_2 injection. When CO_2 comes in contact with brine, the brine is pushed away while vaporized by CO_2. Since the brine is being vaporized while it is moving, the displaced brine saturation will be less than the movable brine saturation. The higher the mobility of the brine, the more brine is displaced and less chance for vaporization. We approximated the three-component (CO_2, water, salt) system with a two-component (CO_2, brine) system to construct the saturation profile. The analytical solution found to be composed of three regions that are connected by two shocks (trailing and leading). The trailing shock velocity was found to be the same as the velocity of the brine saturation downstream. Therefore, all the saturations with velocities lower than the trailing shock velocity will not appear in the solution.

The brine will be fully vaporized once it reaches the saturation downstream of the trailing shock. Then, the gaseous phase which enters the equilibrium region will be already saturated by water.

The leading shock moves forward due to movement of the encountered fresh brine by gaseous phase. The brine behind the leading shock is already saturated by CO_2 since the dissolution occurs at the leading shock only. Therefore, there will be no mass transfer between the saturated aqueous phase behind the leading shock and saturated gaseous phase ahead of the trailing shock. In other words, there will be no salinity change in the equilibrium region. Hence, all the solid salt precipitation will occur behind the trailing shock as a result of vaporizing the saturated brine downstream of the trailing shock ($S_a^c = 1 - S_g^c$). This region is known as the dry-out zone. Static material balance calculations give the equation below for the solid salt saturation in the dry-out region the details of which can be found in [8]:

$$S_s = \frac{s}{\rho_s}\left(\omega_{water,g} S_g^c \rho_g + \omega_{Brine,a}\left(1 - S_g^c\right)\rho_a\right) \qquad \text{or} \qquad S_s = \frac{s\rho_{inj} G_{Brine}^c}{\rho_s} \quad \textbf{(8)}$$

Based on this equation the precipitated salt saturation depends on salinity(s), solid salt density (ρ_s), injected CO_2 density (ρ_{inj}), and the global concentration downstream of the trailing shock (G_{Brine}^c).

6. Porosity and Permeability Change

Using a simple Kozeny-Carman grain model, the ratio of the permeability k to initial permeability k_0 can be expressed as:

$$\frac{k}{k_0} = \left(\frac{\phi}{\phi_0}\right)^3 \left(\frac{1-\phi_0}{1-\phi}\right)^2 \quad \textbf{(9)}$$

where ϕ_0 is the initial porosity. Equations 9 combined with the change in porosity: $\phi = \phi_0(1 - S_s)$ gives:

$$\frac{k}{k_0} = \frac{\left(1-S_s\right)^3}{\left(1 + \frac{\phi_0}{1-\phi_0} S_s\right)^2} \quad \textbf{(10)}$$

Since the solid salt saturation is constant, the permeability reduction in the dry-out zone is independent of radius. However, with the expansion of the dry-out zone, the permeability reduction affects larger area of the aquifer over time. The change in permeability can be described by the skin factor to be included in the reservoir simulation [9]:

$$sf = \left(\frac{\left(1 + \frac{\phi_0}{1-\phi_0} S_s\right)^2}{\left(1-S_s\right)^3} - 1 \right) \ln\left(\frac{r_{dry}(t)}{r_w}\right) \quad \textbf{(11)}$$

where r_{dry} is the radius of the dry-out region which is time-dependent. Therefore, the resulted skin factor is time-dependent.

7. Model Verification

The analytical model described above is now applied to a reference case which is simulated numerically. The analytical solution assumes homogeneity, no diffusion/dispersion, no capillary and gravity forces, non-reactive rock, local phase equilibrium, and constant pressure and temperature. Moreover, the analytical model approximates the ternary system with a binary system in which the salt precipitation only occurs over the dry-our zone. The numerical results are based on totally different formulation that does not have these assumptions or approximations. Comparison between analytical and numerical results can help to investigate the relevancy of our assumptions and approximation.

8. Reference Case

In absence of experimental data the results of the analytical model described in this paper are compared to those obtained by numerical simulations. The analytical model is applied to a basic problem of CO_2 injection into a saline aquifer with simplified flow geometry and aquifer properties. CO_2 is injected into a homogeneous, isotropic, infinite-acting aquifer at conditions of 120 bar pressure, 45 C temperature, and a salinity of 15 wt%. CO_2 is injected at a constant rate of 100 kg/s (3.15 Mt/year) through a well in aquifer centre which is completed over the whole aquifer thickness of 100m. The flow is assumed 1-D radial and gravity effects are neglected. This problem has been presented in the Lawrence Berkeley National Laboratory (LBNL) inter-comparison project [10] as test Problem #3. The numerical results for this problem are presented by Pruess and Spycher [11], using a TOUGH2 simulator with the ability to simulate the salt precipitation. The PVT data used in the numerical work is based on the PVT model presented by Spycher and Pruess [12].

9. Analytical Solution

In development of the analytical solution, the radial CO_2/brine model is used first to locate the shocks and to find the profiles of gaseous phase saturation. The profile of gaseous phase saturation is illustrated in Figure 4. The dry-out region(where salt precipitation occurs) will cover an area of 60 m radius after 10 year of injection in this case. Moreover, 2206 m of the aquifer will be accessible by the CO_2 after this time.

We can now use the salt precipitation formulation to calculate the solid salt saturation in the dry-out zone. The solid salt saturation is calculated to be 0.038 in the dry-out zone. Note that we are using Hassanzadeh et al. PVT model [13] in the analytical solution. This model is in close agreement with that of Spycher and Pruess [12].

10. Comparison of the Analytical Model with the Numerical Results

Pruess and Spycher [11], have shown that the numerical results for the reference case follow a similarity solution, where the solution remains invariant when plotted versus the similarity variable r^2/t (note that the numerical results are given for the line source case, i.e. r_w=0). This is in agreement with the analytical solution developed in this paper. The analytical versus numerical results are shown in Figures 5 and 6 with respect to the similarity variable. The simulation results show variable solid salt saturation in the dry-out zone, which could be the result of numerical instability problems. The numerical model gives a slower leading shock and a faster trailing shock. As the pressure varies in the numerical simulation, the compositional properties change as well. This will cause a difference between the shock specifications of the numerical and analytical solutions. However, the agreement is good enough to establish that the TOUGH2 and the analytical solution are solving the same problem with good agreement.

The salt precipitation occurs behind the trailing shock (dry-out zone) only. Based on Equation 10, the permeability of the dry-out zone reduces to 0.87 of its initial value due to salt precipitation. For this case, the skin factor cannot be calculated using Equation 11, as the well radius (r_w) is taken as zero (line source).

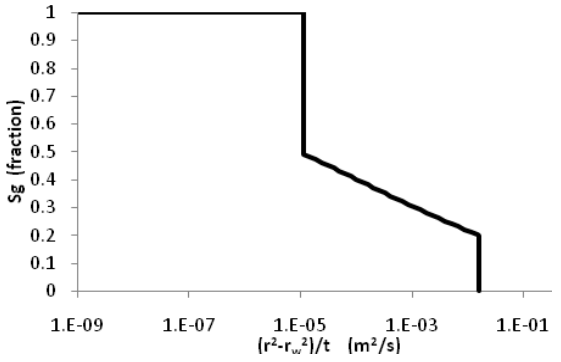

Figure 4: Analytical gaseous phase saturation profile (salt precipitation neglected)

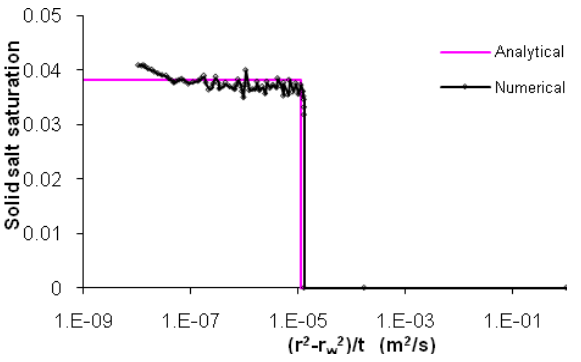

Figure 5: Analytical vs. numerical results for solid saturation

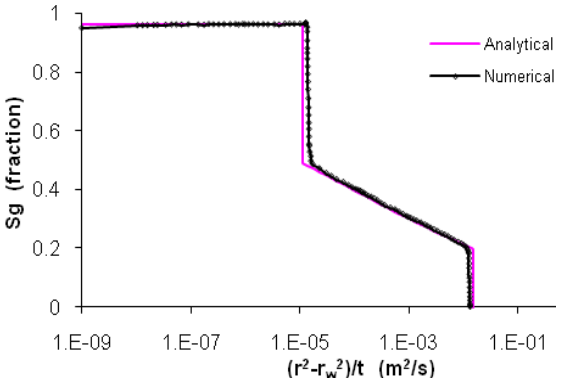

Figure 6: Analytical vs. numerical results for gaseous phase saturation.

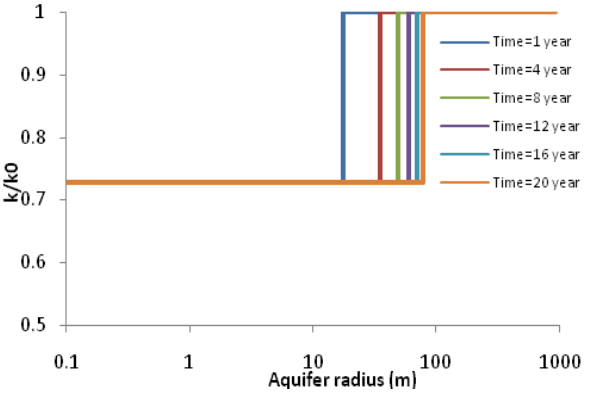

Figure 7: Analytical results for permeability reduction when the brine is fully saturated.

If the analytical model is applied to the problem above with halite-saturated brine (salinity of 30 wt%), the solid salt saturation can be as high as 0.086 in this case. As a result, the permeability reduces up to 0.73 of its initial value due to salt precipitation. Figure 7 shows the ratio of current to initial permeability versus aquifer radius for different times. An area of 100 meters radius will be affected by salt precipitation after 20 years.

11. Summary and Conclusion

Due to salt precipitation, as water evaporates into injected CO_2, porosity and permeability might reduce in some CO_2 injection scenarios. An analytical model is developed to predict the amount of salt precipitation and the region in which it occurs over time. The model assumes homogeneity, no diffusion/dispersion, no gravity and capillary forces, non-reactive rock, local phase equilibrium, and constant pressure and temperature. In development of this model, Orr's formulation on two-component CO_2/brine displacement is translated from a Cartesian to a radial coordinate system at first. Next, a model for salt precipitation is presented and included in the radial 2-phase model. It is shown that the model has a similarity property, which means that the solution depends on radial distance r and time t only through the similarity variable $\eta=(r^2-r_w^2)/t$. The similarity variable η known as wave velocity, is the local flow rate times the fractional flow derivative. This definition of wave velocity gives a physically impossible solution because we find that multiple saturations will exist at a single location. The inconsistency is resolved by the propagation of discontinuities known as shocks. Two shocks are developed as a result: the trailing shock and the leading shock. Approximation of the ternary system by a binary system causes the salt precipitation to occur in the regions behind the trailing shock (dry-out zone) only. The salt precipitation in the dry-out zone occurs due to vaporization of the brine saturation downstream of the trailing shock. The model is extended to evaluate the effect of salt precipitation on porosity and permeability. It is also shown that the effect can be described in terms of time-dependent skin factor.

The analytical model was applied to the LBNL inter-comparison problem #3. The analytical results match the numerical results using the TOUGH2 simulator. The results show that the salt precipitation can be significant, especially when the brine is halite-saturated. In such a case the permeability may reduce to 73 percent of its initial value.

Acknowledgement:

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Nomenclature:

f = fractional flow
G= normalized global concentration, Dimensionless
H= normalized molar rate, Dimensionless
h = aquifer thickness, m
k = permeability, Darcy
q = volumetric rate, m^3/s
r = radius, m
S = saturation
s = salinity, mole fraction
sf = skin factor
t = time, s
ϕ =porosity, fraction
ω =mole fraction
μ=viscosity, Pa.S
ρ= molar density, $kmol/m^3$
η=similarity variable, m^2/s

Subscript:
0 = initial
D = dimensionless
dry= Dry-out region
g = gaseous phase
inj = injection
s = Solid phase
a= aqueous phase
t = total
w = well

Superscript:
b = upstream of the leading shock
c = downstream of the trailing shock

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